

A statics-dynamics equivalence through the fluctuation–dissipation ratio provides a window into the spin-glass phase from nonequilibrium measurements

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We have performed a very accurate computation of the nonequilibrium fluctuation–dissipation ratio for the 3D Edwards–Anderson Ising spin glass, by means of large-scale simulations on the special-purpose computers Janus and Janus II. This ratio (computed for finite times on very large, effectively infinite, systems) is compared with the equilibrium probability distribution of the spin overlap for finite sizes. Our main result is a quantitative statics-dynamics dictionary, which could allow the experimental exploration of important features of the spin-glass phase without requiring uncontrollable extrapolations to infinite times or system sizes.

spin glasses | fluctuation–dissipation relation | glasses | statics-dynamics equivalence | out-of-equilibrium dynamics

Theory and experiment follow apparently diverging paths when studying the glass transition. On the one hand, experimental glass formers (spin glasses, fragile molecular glasses, polymers, colloids, and ...) undergo a dramatic increase of characteristic times when cooled down to their glass temperature, T_g (1). Below T_g , the glass is always out of equilibrium and “aging” appears (2). Consider a rapid quench from a high temperature to the working temperature T ($T < T_g$), where the system is left to equilibrate for time t_w and probed at a later time $t + t_w$. Response functions such as the magnetic susceptibility turn out to depend on t/t_w^μ , with $\mu \approx 1$ (2–4). The age of the glass, t_w , remains the relevant time scale even for t_w as large as several days. Relating the aging experimental responses to equilibrium properties is an open problem.

A promising way to fill the gap is to establish a statics-dynamics dictionary (SDD) (5–8): nonequilibrium properties at “finite times” t , t_w , as obtained on samples of macroscopic size $L \rightarrow \infty$, are quantitatively matched to equilibrium quantities computed on systems of “finite size” L [the SDD is an $L \leftrightarrow (t, t_w)$ correspondence]. Clearly, in order for it to be of any value, an SDD cannot strongly depend on the particular pair of aging and equilibrium quantities that are matched.

Some time ago, we proposed one such a SDD (6–8). However, this SDD was unsatisfactory in two respects. First, L was matched only to t_w (irrespective of the probing time $t + t_w$). Second, our

SDD matched spatial correlation functions whose experimental study is only incipient (9, 10).

One could think (5) of building an SDD through the generalized fluctuation–dissipation relations (GFDRs) first introduced in ref. 11 (for related developments, see refs. 12–19). The GFDRs are correct at very large times. However, on time scales that can be investigated in experiments, glassy systems are not fully thermalized because the approach to equilibrium is very slow. Strong corrections pollute GFDRs at finite times.

Significance

The unifying feature of glass formers (such as polymers, supercooled liquids, colloids, granulars, spin glasses, superconductors, etc.) is a sluggish dynamics at low temperatures. Indeed, their dynamics are so slow that thermal equilibrium is never reached in macroscopic samples: in analogy with living beings, glasses are said to age. Here, we show how to relate experimentally relevant quantities with the experimentally unreachable low-temperature equilibrium phase. This relation is made quantitative via a statics-dynamics dictionary, established for spin glasses. In our dictionary, the aging response to a magnetic field is related to the spin-glass order parameter as obtained on samples small enough to equilibrate. We remark that all of the observables we consider can be measured with current experimental methods.

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Here we show how the SDD can be used in a particular case to compute such corrections (that will be likely present in all glassy systems). We find that the naive implementation of this idea (5) does not work in general, and we introduce a modified SDD that works for spin glasses (and, hopefully, also for glasses).

GFDRs carry crucial information (11, 14, 15): they provide a promising experimental path toward measuring Parisi's functional order parameter (20). As a consequence, GFDRs have attracted much attention. One encounters numerical studies for both Ising (13, 16, 18) and Heisenberg (21, 22) spin glasses, as well as for structural glasses (23–27). On the experimental side, we have studies on atomic spin glasses (17, 19), superspin glasses (10), polymers (9, 28), colloids (29–35) or DNA (36).

Here, we perform a detailed simulation of GFDRs in the 3D Ising spin glass using the custom-made supercomputers Janus (37) and Janus II (38). In fact, this study has been the launching simulation campaign of the Janus II machine, which was designed with this sort of dynamical studies in mind. Our simulations stand out by the spanned time range (11 orders of magnitude), by our high statistical accuracy and by the range of system sizes, enabling us to control size effects ($L = 20, 40, 80$ and 160). Thus, armed, we assess whether or not an SDD can be built from the GFDR and compare the SDD proposed in this paper with other proposals. We focus on spin glasses, rather than on other model glasses, for a number of reasons: (i) their sluggish dynamics is known to be due to a thermodynamic phase transition at $T_c = T_g$ (39–41); (ii) the linear size of the magnetically correlated domains, $\xi(t_w)$, is experimentally accessible (42, 43) ($\xi \sim 100$ lattice spacings (42), much larger than comparable measurements for structural glasses (44)); (iii) a GFDR-based SDD has been well established in the limit of large sizes and times (11, 14, 15) (Eq. 4); (iv) GFDRs have been studied experimentally (17); (v) well-developed, yet mutually contrasting, theoretical scenarios are available for spin glasses in equilibrium (45); (vi) magnetic systems are notably easier to model and to simulate numerically [in fact, special-purpose computers have been built for the simulation of spin glasses (37, 38, 46–48)].

Results

GFDRs and the SDD. We suddenly cool a 3D spin-glass sample of size L^3 from high temperature to the working (subcritical) temperature $T = 0.7 = 0.64T_c$ at the initial time $t_w = 0$ (see *Materials and Methods* for more details and definitions). During the nonequilibrium relaxation a coherence length $\xi(t_w)$ grows (6, 42, 49), which is representative of the size of the spin-glass domains. Then, from the waiting time t_w on, we place the system under a magnetic field of strength H , and consider the response function at a later measuring time $t + t_w$

$$\chi_L(t + t_w, t_w) = \left. \frac{\partial m_L(t + t_w)}{\partial H} \right|_{H=0}, \quad [1]$$

where $m_L(t + t_w)$ is the magnetization density in a sample of linear size L . This susceptibility is then compared with the spin-temporal correlation function $C_L(t + t_w, t_w)$. From now on, we shall take the limits

$$\chi(t + t_w, t_w) = \lim_{L \rightarrow \infty} \chi_L(t + t_w, t_w), \quad [2]$$

$$C(t + t_w, t_w) = \lim_{L \rightarrow \infty} C_L(t + t_w, t_w), \quad [3]$$

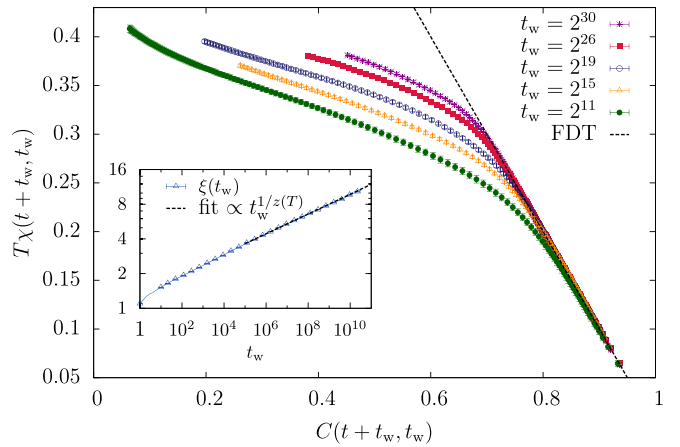


Fig. 1. Response function $T\chi(t + t_w, t_w)$ versus $C(t + t_w, t_w)$ at $T = 0.7$ [for fixed t_w , $C(t + t_w, t_w)$ monotonically decreases from $C = 1$ at $t = 0$ to $C = 0$ at $t = \infty$]. Data for $t_w = 2^{11}$ and $t_w = 2^{30}$ were obtained on Janus II (the other t_w are from Janus). The five values of t_w correspond to effective equilibrium sizes L_{eff} that, according to Eq. 6, span the size range investigated in ref. 7 (namely, $8 \leq L \leq 32$). (Inset) Growth of the spin-glass coherence length $\xi(t_w)$ as a function of time, computed at zero magnetic field and following refs. 6 and 49, from simulations of $L = 160$ lattices at $T = 0.7$ on Janus II. In dashed lines, we plot the scaling $\xi(t_w) \propto t_w^{1/z(T)}$ with $z(T) = 11.64$ from ref. 48.

which are easy to control numerically: if $L \gtrsim 7\xi(t + t_w)$ size effects are negligible (6)* (also see *SI Appendix*).

The Fluctuation–Dissipation Theorem (FDT) states that $T\chi(t + t_w, t_w) = 1 - C(t + t_w, t_w)$, with both χ and C computed at $H = 0$. However, for $T < T_c$ the FDT does not hold. In fact, GFDRs take the form (11, 14, 15) (the order of limits is crucial):

$$\lim_{t_w \rightarrow \infty} T\chi(t + t_w, t_w) = \lim_{t_w \rightarrow \infty} [\lim_{L \rightarrow \infty} S(C_L(t + t_w, t_w), L)], \quad [4]$$

where t is scaled as t_w grows, to ensure that the full range $0 < C(t + t_w, t_w) < 1$ gets covered, and $S(C, L)$ is given by a double integral of $P(q, L)$, the equilibrium distribution function of the spin overlap, whose explicit definition is provided in *Materials and Methods*.

Here, we mimic an experimental protocol (17, 19) in that we consider the nonequilibrium response on a very large system but at finite times. We try to relate this response with the equilibrium overlap for a system of finite effective size L_{eff}

$$T\chi(t + t_w, t_w) = S(C(t + t_w, t_w), L_{\text{eff}}(t + t_w, t_w)), \quad [5]$$

where we have assumed that both χ and C have reached their thermodynamic limit. The same approach was followed for a 2D spin glass by Barrat and Berthier (5) (note, however, that there is no stable spin-glass phase at $T > 0$ in two spatial dimensions).

Eq. 5 provides a SDD relating both times t and t_w with a single effective equilibrium size $L_{\text{eff}}(t + t_w, t_w)$. Note that it is not obvious a priori that our program can be carried out. For instance, our SDD does not exist for ferromagnets, as explained in detail in the *SI Appendix*, using data from refs. 50 and 51.

SDDs based on the comparison of aging and equilibrium correlation functions (rather than on GFDRs) have been studied in some detail (7, 8, 52). It was found that the effective length depends solely on t_w . Indeed,

$$L_{\text{eff}}(t + t_w, t_w) = k\xi(t_w), \quad [6]$$

*In fact, the correlation functions decay exponentially with distance. Therefore, with periodic boundary conditions, size effects should decay exponentially with L/ξ . Indeed, an explicit computation shows that, to our accuracy level, size corrections are completely negligible when $L > 7\xi$ (6).

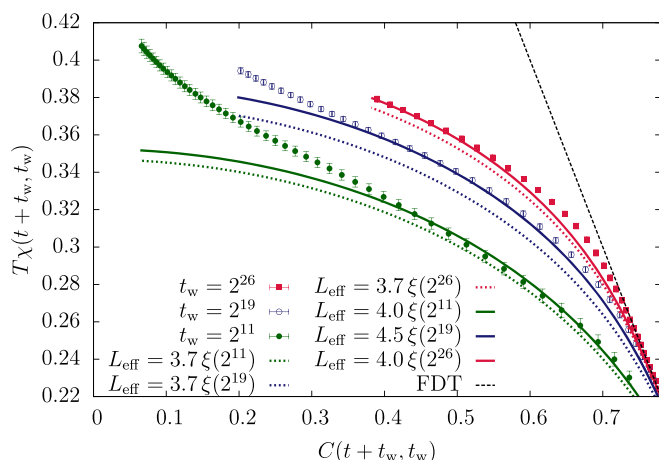


Fig. 2. Close-up of Fig. 1. (We only show data for three t_w , for the sake of clarity.) Lines are $S(C, L_{\text{eff}})$; recall Eq. 5, with the effective equilibrium size as in Eq. 6: $L_{\text{eff}}(t + t_w, t_w) = k\xi(t_w)$. Dotted lines correspond to $k = 3.7$, which is the proportionality constant that was found by matching equilibrium and nonequilibrium correlation functions (6–8). The continuous lines were found by choosing the best possible k for each t_w . This representation shows that the single-time SDD $L_{\text{eff}} \sim \xi(t_w)$ breaks down for large t , when $\xi(t + t_w)$ is much larger than $\xi(t_w)$.

with $k \approx 3.7$, was accurate enough to match the correlation functions (7, 8). Ref. 5 also agreed with Eq. 6. In fact, Eq. 6 also underlies the analysis of refs. 53 and 54. However, we shall show below that Eq. 6 is oversimplified.

Numerical Data. The three basic quantities computed in this work, namely $\chi(t + t_w, t_w)$, $C(t + t_w, t_w)$, and $\xi(t_w)$, are displayed in Fig. 1. Full details about this computation are provided in [SI Appendix](#).

Let us remark that the Janus II supercomputer allows us to probe unexplored dynamical regimes, either t/t_w as large as $2^{24} \approx 1.4 \times 10^7$ (i.e., we follow the magnetic response for a very long time, after the field was switched on at $t_w = 2^{11}$) or t_w as large as 2^{30} (i.e., we study the response of a very old spin glass, but we are limited to $t/t_w \approx 27$ in this case).

It is also remarkable that we are able to compute both the susceptibility χ and the correlation function C without worrying about finite-size effects. Indeed, size effects become visible when the coherence length reaches the threshold $\xi(t_w) \approx L/7$ (6) which in our $L = 160$ lattice translates to $\xi \approx 23$ lattice spacings. As Fig. 1, *Inset* shows, we are quite far from this safety threshold.

With respect to previous measurements of the GFDR ratio, it is worth stressing that now we are able to take the $h \rightarrow 0$ limit in a more controlled way. This extrapolation is far from trivial, given that the linear response regime shrinks to very small field when t_w increases (*SI Appendix*).

The data in Fig. 1 also stand out by their statistical accuracy (due to the large number of samples and large system sizes we simulated, but also thanks to the analysis method described in *SI Appendix*). As a consequence, a behavior different from the one implied by FDT, $T\chi(t, t_w) = 1 - C(t, t_w)$, can be studied in detail. In particular, the reader might be stricken by the linear behavior at $C(t + t_w, t_w) \approx 0.4$. In fact, following refs. 11, 14, and 15, this linear behavior could be interpreted as evidence for one step of replica-symmetry breaking (see, for instance, ref. 55). However, we shall argue below that the effective length in Eq. 5 evolves as time t grows, thus producing an upturn in the response which is probably responsible for the linear behavior in Fig. 1.

Let us make a final remark. We know that $S(C, L)$ is upper bounded by $1 - \overline{\langle |q| \rangle}_{L=\infty} \geq 1 - q_{\text{EA}}^{(L=\infty)}$ (see *Materials and Methods* for definitions; the proof of the inequality is outlined in [SI](#)

Appendix). At $T = 0.7$ we know that $1 - q_{\text{EA}}^{(L=\infty)} = 0.48(3)$ (8) [or $0.46(3)$ (7)]. Therefore, the dynamic responses $T\chi(t, t_w)$ in Fig. 1 are well below $1 - q_{\text{EA}}^{(L=\infty)}$ and Eq. 5 could be satisfied. The general conditions under which Eq. 5 can be used are discussed in *SI Appendix*.

The Effective Equilibrium Size. As we show in Fig. 2, our data are too accurate to be quantitatively described by combining Eq. 5 with Eq. 6. This simple description fails both at short times t (i.e., when $C(t, t_w) \approx q_{\text{EA}}^{[L \approx 4\xi(t_w)]}$) and also at very long t , although one can find a constant k that works well for intermediate t .

The discrepancy for long t seems easy to rationalize: because the growth of $\xi(t_w)$ is very slow (recall Fig. 1, *Inset*) $\xi(t + t_w)$ and $\xi(t_w)$ are very similar to each other for small t and, therefore, $L_{\text{eff}} \propto \xi(t_w)$ makes sense. However, because $\xi(t_w)$ grows without bounds in the spin-glass phase, one should eventually have $\xi(t + t_w) \gg \xi(t_w)$. Under these circumstances, it is only natural that $L_{\text{eff}} \propto \xi(t + t_w)$.

We can test this proposal by computing an exact L_{eff} for each (t, t_w) pair (see *SI Appendix* for details), which we plot in Fig. 3: in the main graph in units of $\xi(t + t_w)$ and in the inset in units of $\xi(t_w)$.

The first important observation from the main panel in Fig. 3 is that, for long enough times, we find $L_{\text{eff}} \approx 2.6 \xi(t + t_w)$, in agreement with the intuition exposed above. This SDD is definitely different from Eq. 6, used until now. The data in Fig. 3, *Inset* explain why the previous relation in Eq. 6 passed many numerical tests until now: the nonmonotonic behavior of $L_{\text{eff}}/\xi(t_w)$ for short times t makes this ratio roughly compatible with a constant $k \approx 4$ as long as $t/t_w \lesssim 1000$.

Surprisingly, the ratio $L_{\text{eff}}/\xi(t + t_w)$, or equivalently $L_{\text{eff}}/\xi(t_w)$, becomes large as well when $t \rightarrow 0$, thus explaining the inability of Eq. 5 in describing dynamical data at short times t (Fig. 2). Nonetheless in the limit $t \rightarrow 0$, i.e., $\xi(t + t_w)/\xi(t_w) \rightarrow 1$, the effective equilibrium size L_{eff} seems to reach a finite value; a divergence of L_{eff} in this limit seems unlikely (*SI Appendix*).

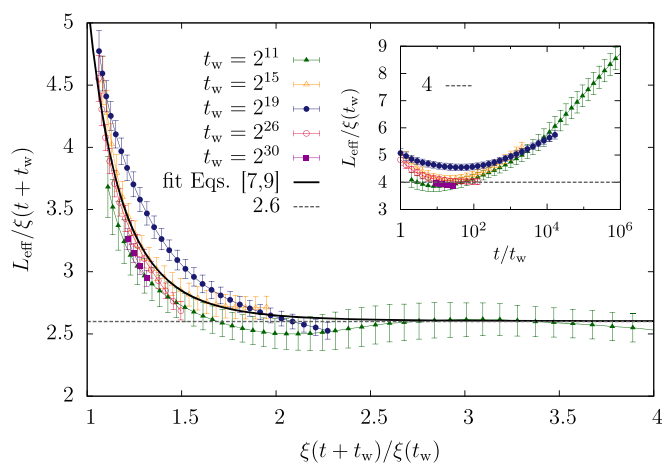


Fig. 3. For each t_w , we show the effective equilibrium size $L_{\text{eff}}(t + t_w, t_w)$ in units of the coherence length at the measuring time $\xi(t + t_w)$ versus the ratio of coherence lengths $\xi(t + t_w)/\xi(t_w)$ (recall that t is the time elapsed since switching-on the magnetic field). The ratio of coherence lengths is 1 for $t = 0$ and goes as $\xi(t + t_w)/\xi(t_w) \propto (1 + t/t_w)^{1/z(T)}$ for large time, with $z(T = 0.7) = 11.64(15)$ (49). Let us stress that there is no extrapolation in this figure, only interpolation (i.e., L_{eff} falls within the simulated equilibrium sizes, $8 \leq L_{\text{eff}} \leq 32$). The solid line is a fit to the scaling function $h(x)$ in Eqs. 7 and 9. (*Inset*) $L_{\text{eff}}(t + t_w, t_w)$ data from the main panel in units of the coherence length at the initial time $\xi(t_w)$, as a function of the time ratio t/t_w .

The outcome of this analysis is given in Fig. 5. It turns out that the simplified S_{simpl} in Eq. 11 is almost as effective as the true $S(C, L)$ in representing the nonequilibrium data through the effective size L_{eff} in Eq. 9. The only obvious disagreement is that Eq. 11 predicts a nonanalytic behavior for the susceptibility χ at $C = q_{\text{EA}}^{(L_{\text{eff}})}$, which is not found in the nonequilibrium data. In other words, the effective size for times such that $C(t + t_w, t_w) \approx q_{\text{EA}}^{(L \approx 4\xi(t_w))}$ is large, but certainly L_{eff} is not infinite as demanded by Eq. 10.

Fortunately, even the crude description in Eq. 11 could lead to some interesting analysis. For instance, one could select pairs of times (t, t_w) such that $L_{\text{eff}}(t + t_w, t_w) = \text{constant}$. Then, $S(0, L_{\text{eff}})$ will be the same for all those points. Now, we note from Eq. 9 that $\xi(t + t_w)$ can vary by as much as a factor of two, for such points. It follows that $C(t + t_w, t_w)$ should vary significantly over this set of times with fixed $L_{\text{eff}}(t + t_w, t_w)$. Hence, the crucial parameters P_0 and P_1 could be extracted. For instance, if the susceptibility $\chi(t, t_w)$ turned out not to depend on $C(t + t_w, t_w)$ for fixed L_{eff} , then we would have $P_0, P_1 \approx 0$, in contrast with the mean field prediction $P_0 > 0$.

Discussion

It was discovered some twenty years ago that experimental aging response functions carry information on Parisi's functional order parameter (11–13). We now know that this connection between nonequilibrium and equilibrium physics relies on a very general mathematical property, stochastic stability (14, 15), shared by many glass models. However, experimental attempts to explore this connection encountered a major problem (17, 19): an essentially uncontrolled extrapolation to infinite waiting time t_w is required. (See ref. 57 for an experimental attempt to measure Parisi's functional order parameter, unrelated to GFDRs.)

Here, we have proposed using a SDD (5–8) to avoid uncontrolled extrapolations. Indeed, we have shown that the aging responses at finite t_w can be connected to the Parisi's order parameter as computed at equilibrium in a system of finite size.

We have shown that this GFDR-based SDD is essentially consistent with previous proposals (6–8) that focused on spatial correlation functions. This is an important consistency test. There is a caveat, though: when the probing time $t + t_w$ is such that one has $\xi(t + t_w) \gg \xi(t_w)$ for the coherence lengths, the GFDR-based SDD disagrees from previous dictionaries in that the size of the equivalent equilibrium system is $L_{\text{eff}} \sim \xi(t + t_w)$ [rather than $L_{\text{eff}} \sim \xi(t_w)$]. In fact, we have found that the L_{eff} dependence on both length scales can be simply parameterized, recall Eqs. 7 and 9.

At this point, the reader may wonder about the relationship between $L_{\text{eff}}(t + t_w, t_w)$ and the two-time correlation length $\zeta(t + t_w, t_w)$ obtained from the two-time-two-site correlation function introduced in refs. 58 and 59. Indeed, we thoroughly studied the two-time/two-site correlation function in ref. 49 because it was a crucial ingredient for our previous SDD proposal (7, 8). We found (figure 12 in ref. 49) that $\zeta(t + t_w, t_w)$ can grow, at most, as large as $\xi(t_w)$. Instead, the $L_{\text{eff}}(t + t_w, t_w)$ introduced here is asymptotically as large as $\xi(t + t_w)$.

On the other hand, the only previous SDD known to us that was based on Eq. 5 misses the $L_{\text{eff}} \sim \xi(t + t_w)$ behavior (5). There are a couple of possible reasons for this failure. For one, the time scales in ref. 5 do not allow for length-scale separation $\xi(t + t_w) \gg \xi(t_w)$. Besides, the SDD from ref. 5 was obtained for 2D spin glasses (which only have a paramagnetic phase). Therefore, the results of ref. 5 are probably a manifestation of finite-time/finite-size scaling (52, 60).

Let us conclude by stressing that the three basic quantities analyzed in this work, namely the susceptibility $\chi(t + t_w, t_w)$, the correlation function $C(t + t_w, t_w)$ and the coherence length $\xi(t + t_w)$, have been obtained experimentally in a dynamic set-

ting very similar to simulations (for χ and C , see refs. 17 and 19; for ξ , see refs. 42 and 43). We thus think that it should be possible to extract the spin-glass functional order parameter from already existing experimental data. Furthermore, GFDRs have been studied as well in superspin glasses (10) and in a variety of soft condensed-matter systems (9, 28–36). We therefore expect that our analysis will be of interest beyond the realm of spin glasses.

Materials and Methods

We study the $D=3$ Edwards–Anderson model, whose Hamiltonian is given by

$$\mathcal{H} = - \sum_{\langle x,y \rangle} J_{x,y} \sigma_x \sigma_y - H \sum_x \sigma_x. \quad [12]$$

The spins $s_x = \pm 1$ are placed on the nodes, x , of a cubic lattice of linear size L , and we set periodic boundary conditions. The couplings $J_{x,y} = \pm 1$, which join nearest neighbors only, are chosen randomly with 50% probability and are quenched variables. For each choice of the couplings (one “sample”), we simulate two independent copies of the system, $\{s_x^{(1)}\}$ and $\{s_x^{(2)}\}$. We denote by $\langle \dots \rangle$ the average over the thermal noise and by $\langle \dots \rangle$ the subsequent average over the samples. The model described by Eq. 12 undergoes a SG transition at $H = 0$ and $T_c = 1.102(3)$ (61).

For our dynamical data, we have run new nonequilibrium simulations on Memento, Janus and Janus II. We use heat-bath dynamics, in which one Monte Carlo step roughly corresponds to one picosecond of the experimental system (62). See SI Appendix for technical details of these simulations. The two main dynamical observables are the magnetization density $m_L(t + t_w) = \overline{\sum_x \langle s_x(t + t_w) \rangle} / V$ and the spin-temporal correlation function $C_L(t + t_w, t_w; H) = \overline{\sum_x \langle s_x(t_w) s_x(t + t_w) \rangle} / V$.

Equilibrium results at $T = 0.7$ are available for $L \leq 8 \leq 32$ (7). In this case the main quantity is the probability density function $P(q, L)$ of the spin overlap q :

$$q \equiv \frac{1}{V} \sum_x s_x^{(1)} s_x^{(2)}, \quad \overline{(q^k)}_L = \int_{-1}^1 dq' (q')^k P(q', L). \quad [13]$$

In particular, we are interested in the integral

$$S(C, L) = \int_C^1 dC' x(C', L), \quad x(C, L) = \int_0^C dq 2P(q, L). \quad [14]$$

The $P(q, L)$ curves are easily described for finite L . They are symmetric under $q \leftrightarrow -q$, with two maxima at $\pm q_{\text{EA}}^{(L)}$ and a flat central region. In the thermodynamic limit, the two peaks turn into delta functions at $\pm q_{\text{EA}}^{(\infty)}$, which mark the maximum possible value of $|q|$. The size evolutions, as checked for $L \leq 32$ (7), are as follows: $q_{\text{EA}}^{(L)} - q_{\text{EA}}^{(\infty)} \propto L^{-\theta \approx 0.38}$ [at $T = 0.7$, $q_{\text{EA}}^{(\infty)} = 0.52(3)$ (8)], the width of the peaks at $\pm q_{\text{EA}}^{(L)}$ scales as $L^{-\beta \approx 0.28}$ while $P(q = 0, L)$ turns out to be greater than zero and L -independent.

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